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***Non-invasive acoustic monitoring of D2O concentration***☐ SP-1☐ INSEP Action Sheet: # [NN], with [Country]**24.1.3.1 Nondestructive Assay (NDA) Technology****Abstract**

*There is an urgent need for real-time monitoring of the hydrogen /deuterium ratio (H/D) for heavy water production monitoring. Based upon published literature, sound speed is sensitive to the deuterium content of heavy water and can be measured using existing acoustic methods to determine the deuterium concentration in heavy water solutions. We plan to adapt existing non-invasive acoustic techniques (Swept-Frequency Acoustic Interferometry and Gaussian-pulse acoustic technique) for the purpose of quantifying H/D ratios in solution. A successful demonstration will provide an easily implemented, low cost, and non-invasive method for remote and unattended H/D ratio measurements with a resolution of less than 0.2% vol.*

**Mission Relevance and Associated Work**

Current methods for H/D determination require periodic sampling and analysis. This approach does not provide the opportunity for persistent monitoring and verification by the IAEA and is relatively expensive and inefficient compared to the potential implementation of real-time, online verification by acoustic methods.

According to the IAEA Department of Safeguards Long Term R&D Plan, 2012-2023 (LTRD; STR-375), the following capability and milestone are addressed:

LTRD Capability 5 – Ability to deploy equipment at facilities to meet safeguards requirements.  
LTRD Milestone 5.6 –Develop improved NDA instruments and techniques to address verification of waste and scrap nuclear material with impure composition or heterogeneous isotopic composition.

The objective addressed in the corresponding IAEA Development and Implementation Support Program for Nuclear Verification 2016-2017 (SGTS; STR-382):

SGTS-001, NDA Techniques, Objective 3 – Develop improved instruments and techniques to address verification of waste and scrap nuclear material with impure composition or heterogeneous isotopic composition.

**Associated Work**

1. Non-invasive acoustic-based monitoring of uranium and deuterium in solution  
~ demonstration project for acoustic-based systems development ~  
Funding agency: Pathfinder, Feynman Center for Innovation, Los Alamos National Laboratory  
Project duration: 1 year
2. Multipurpose Acoustic Sensor for Downhole Fluid Monitoring  
~ development of acoustic-based system for fluid properties determination in geothermal wells ~  
Funding agency: DOE-EERE-ARRA (Department of Energy, Office of Energy Efficiency & Renewable Energy, American Recovery and Reinvestment Act of 2009)

Project duration: 3 years

3. Multiphase Flow Meter

~ development of instrumentation/method for oil-gas-water metering in oil wells ~

Funding Agency: Chevron

Project duration: 8 years.

## **Scope of Work**

### **1. Overview of proposed project**

Current methods for H/D determination require periodic sampling and analysis. This approach does not provide the opportunity for persistent monitoring and verification by the IAEA and is relatively expensive and inefficient compared to the potential implementation of real-time, online verification by acoustic methods. Acoustic methods may provide a simpler solution, and can lead to better resolution. Additionally, these methods require only modest electronics (already demonstrated for other metering applications) and reliable electrical connections. This is a significant enhancement over electro-manometer techniques.

We propose an approach that can lead to a precision and accuracy of better than  $\pm 0.2\%$ , volumetric. A quick literature search leads to precisions of  $\pm 0.2$ - $0.4\%$  using other methods [1]. However, all previous methods require drawing of a sample, elaborate sample preparation, are time consuming, and depend on user interpretation. Our approach consists of a clamp-on type device that can accurately measure  $D_2O$  concentration in real-time in real settings.

The underlying physical basis of the sensing technique proposed here is Swept Frequency Acoustic Interferometry (SFAI) [2], and an in-house Gaussian-pulse acoustic technique. We propose to adapt these acoustics techniques to extract accurate sound speed of  $D_2O$  in solutions. Sound speed can easily be related to concentration. The SFAI technique involves the determination of the frequency response of a solid container and the fluid inside it over a wide frequency range. This spectrum consists of a series of regularly spaced resonance peaks that originate from standing waves set up in the fluid-filled sensor cavity. The sound speed is determined from the frequency spacing between any two consecutive resonance peaks. Additionally, a more complex data analysis can provide other fluid properties, including fluid composition, viscosity, and density.

Limited internal funding from LANL (Los Alamos National Laboratory) was provided earlier this year, in order to provide a demonstration of the ability to accurately determine H/D ratio in solutions. While work for this project still continues, we were able to collect a series of data for several concentrations of  $D_2O$  in solutions, for temperatures ranging from 5 -

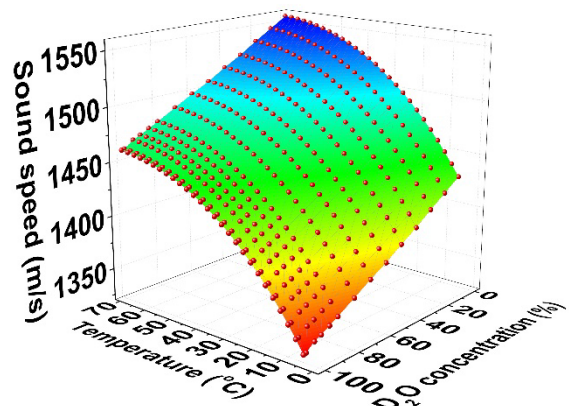


Figure 1. Graphical representation of sound speed, vs temperature and  $D_2O$  concentration.

70°C, for calibration purposes. A 3-dimensional graph of the measured sound speed vs temperature and D<sub>2</sub>O concentration is shown in Figure 1, covering the range of 0 % - 100 % D<sub>2</sub>O (volumetric). The difference in sound speed between pure D<sub>2</sub>O and pure H<sub>2</sub>O is approximately 100 m/s; it varies between 100.5 m/s at 5°C and 94.4 m/s at 70°C. The sound speed can be measured very precisely and accurately, to the first decimal point, resulting in a high precision/accuracy for D<sub>2</sub>O concentration, ~ 0.1% (relative) in laboratory settings. The temperature has to be measured precisely, and variations of temperature in the sample have to be kept to a minimum. At low temperatures (5°C), the temperature has to be measured within 0.03°C. At high temperatures (70°C), measurements within 0.5°C will suffice. Based on previous devices developed in our lab for sound speed measurements, it is expected that similar accuracy and precision can be obtained in real settings.

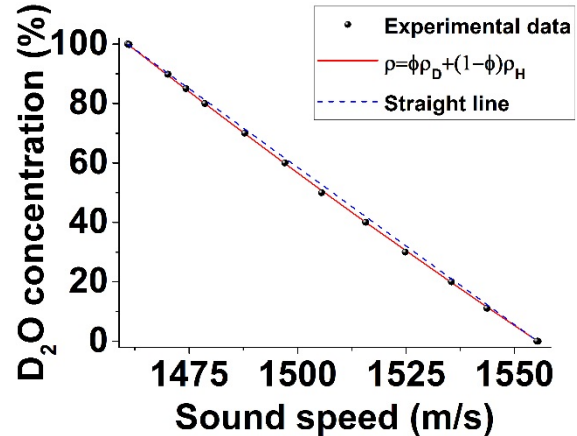


Figure 2. D<sub>2</sub>O concentration vs. sound speed at a temperature of 70°C.

To a first approximation, the D<sub>2</sub>O concentration dependence on sound speed seems to follow a simple linear combination based on the volume fraction  $\phi$ :  $\rho = \phi\rho_D + (1-\phi)\rho_H$ , where  $\rho_D$  is the density of pure D<sub>2</sub>O, and  $\rho_H$  the density of pure H<sub>2</sub>O, see Figure 2, at a temperature of 70°C. The blue dotted line is drawn to guide the eye.

Solution concentrations will be determined from acoustic measurements of the fluid sound speed. These values will be compared with the ones obtained using traditional analytical techniques. At the end of the project we expect to have tested a prototype of the H/D non-invasive acoustic-based system in an actual environment and on different geometries, e.g. pipe – rectangular/cylindrical, either with static liquid or with flow, drum, large tank, etc. The H/D monitoring system is expected to meet IAEA monitoring requirements.

## 2. Description of project trajectory events

We envision this work as a 3-year project. The work is divided in two different concepts of operations (CONOPS). One is (1) a portable system for onsite inspection, while the other is (2) a fixed dual-use system for unattended monitoring/verification.

### Task 1 Portable on-site inspection tool development – CONOPS 1 (FY18-FY19)

Adapt acoustics-based system for clamp-on/bolt-on configurations on realistic storage forms, e.g. process pipes, drums, and/or tank walls, with dimensions and geometries found in real settings, potentially based on NA-241 or IAEA input.

**Deliverable 1:** Portable tool functionality demonstrated in the laboratory (end of FY18).

### Task 2 User-friendly software interface development (FY18)

Develop a user-friendly and simple software interface that is easy to operate by a nontechnical operator.

### Task 3 Continuous unattended monitoring development – CONOPS 2 (FY19)

Refine and adapt instrumentation and software for continuous unattended monitoring that requires minimal input from user/operator.

**Deliverable 2:** Continuous unattended tool functionality demonstrated in the laboratory (end of FY19).

### Task 4 Field tests and technique refinement (FY20)

Test/demonstrate developed system in an actual environment. We are targeting two different sites: (1) the storage facility at Savannah River, and (2) the heavy water plant in Drobeta-Turnu Severin, Romania.

**Deliverable 3:** *Demonstration of system in field functionality similar to the one observed in tests in the laboratory.*

### Pertinent references

1. U.S. Atomic Energy Commission. Laboratory, N.B., N.J. and C.J. Rodden, *Analysis of essential nuclear reactor materials*. 1964: For sale by the Superintendent of Documents, U. S. Govt. Print. Off.
2. Sinha, D., *Noninvasive determination of sound speed and attenuation in chemical warfare agents, in Handbook of Elastic Properties of Solids, Liquids and Gases, vol 4, p 3-21*. 2001: Academic.

### Technology Readiness Level

At the beginning of FY18 the TRL of the proposed technology will be at TRL 5, with tests on a representative model in a somewhat realistic environment. As the project progresses, we will move quickly to a TRL 6 (in the first half of FY18), testing a representative system in a relevant environment, advancing to a TRL 7 in the second year (FY19) – prototype development and tests in an actual environment.

**Technology Readiness Level Table**

Start TRL	End TRL	Component(s)	Notes
TRL5	TRL8	Tool development	System completed and demonstrated through testing and operation in actual conditions.
TRL5	TRL8	OVERALL TRL	Major contributing component: adaptation of existing, established acoustic technique for accurate determination of D <sub>2</sub> O concentration in solutions.

### Summary Table

Task No.	Event Type	Event Title	Responsible Lab	Event Date
1	Milestone	Adapt system for relevant pipes	LANL	09/30/2019
3	Milestone	Unattended monitoring system	LANL	09/30/2019
	Deliverable	Refine system for continuous unattended monitoring	LANL	09/30/2019
4	Milestone	Field tests and system refinement	LANL	09/30/2020
	Deliverable	System in field functionality similar to the one observed in tests in the laboratory	LANL	09/30/2020
	Field Test Plan	Demonstrate system functionality in real conditions	LANL	09/30/2020
	Field Test	Savannah River and Romania	LANL	09/30/2020

**Participating Laboratories**

<b>Lab</b>	<b>Program Manager</b>	<b>Principal Investigator</b>	<b>PI's Email</b>	<b>PI's Phone</b>
LANL*	Rollin Lakis	Cristian Pantea	pantea@lanl.gov	505-665-7598

\*Lead Laboratory

<b>Task</b>	<b>Laboratory</b>	<b>Funding (\$K)</b>
1	LANL	310
2	LANL	185
		Total: 495

**Anticipated Future Needs and Activities**

The final outcome of this project is a product that can be used for IAEA verifications in the field.

<b>Year</b>	<b>Funding (\$K)</b>	<b>Anticipated Future Tasks</b>
<b>FY19</b>	490	CONOPS 2 (unattended verification) development
<b>FY20</b>	475	Field tests and system refinement



# Non-invasive acoustic monitoring of D<sub>2</sub>O concentration



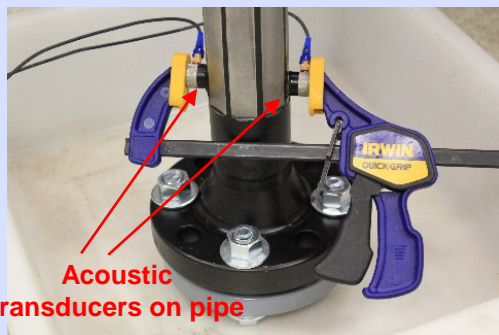
Field portable and process monitoring solutions for heavy water verification

## Background/State of the Art



- Current methods: periodic sampling or invasive continuous monitoring
- No persistent monitoring and verification
- Relatively expensive
- Needs significant user interaction
- No other entity works on acoustics approach

## Innovation

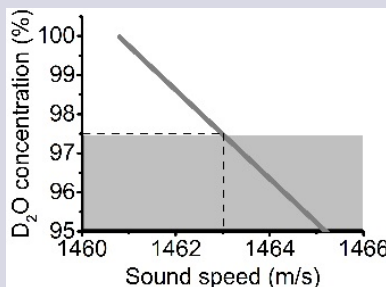
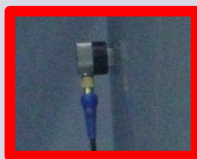


- Use acoustics and clamp-on transducers
- **Noninvasive, unattended, continuous monitoring**
- Preliminary data very promising. Sound speed sensitive to H/D content.

## Approach, Metrics and Outcomes

### MAIN GOAL

- Reduce inspectors presence/increase verification coverage
- Demonstrate functionality in the field on different storage forms (process pipes, drums, tank walls, etc.)

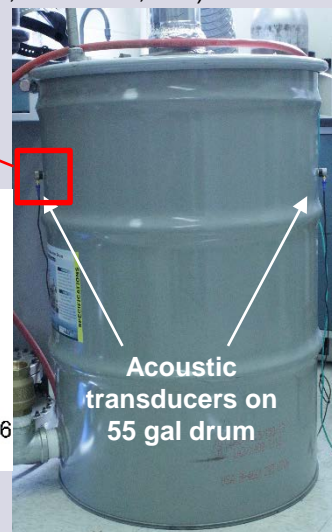


### HOW IT WORKS

- Determine accurate sound speed in fluid using Swept Frequency Acoustic Interferometry (SFAI).
- We already demonstrated high precision/accuracy for D<sub>2</sub>O concentration, ~ 0.1% (relative) in laboratory.

### ASSUMPTIONS, LIMITATIONS & CONSTRAINTS

- Constraints: at low temperatures (5°C), the temperature has to be measured within 0.03°C. However, at high temperatures (70°C), measurements within 0.5°C will suffice.



## Impact

- Safeguards relevance
  - Current approaches do not provide noninvasive continuous monitoring and verification by the IAEA
  - CONOPS 1: man portable tool
  - CONOPS 2: continuous unattended verification
- Long-Term R&D STR-375 LTRD Capability 5/LTRD Milestone 5.6
- IAEA STR-382 Objective: SGTS-001, NDA Techniques, Objective 3
- **Start of FY TRL = TRL5**
- **End of FY TRL (Planned) = TRL6**
- **End of project TRL (Planned) = TRL8**

## Goals/Action Plan

- **Planned tasks:**
  - 1 - Portable on-site inspection tool
  - 2 - User-friendly software interface
  - 3 - Continuous unattended monitoring
  - 4 - Field tests and technique refinement

## Future FY

- Continuous unattended monitoring
- Field tests and technique refinement

## Team

**Los Alamos National Laboratory**

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# NON-INVASIVE ACOUSTIC MONITORING OF D<sub>2</sub>O CONCENTRATION



OFFICE OF  
NONPROLIFERATION AND  
ARMS CONTROL (NPAC)



INTERNATIONAL NUCLEAR SAFEGUARDS

## Point of Contact:

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## Benefit

- Noninvasive, unattended, continuous monitoring. Sound speed very sensitive to H/D content (0.1%).
- Long-Term R&D goals:
  - (1) Reduce inspectors presence/increase verification coverage
  - (2) Demonstrate functionality in the field on different storage forms (process pipes, drums, tank walls, etc.)

## Applications

### Safeguards relevance:

- Current approaches do not provide noninvasive continuous monitoring and verification by the IAEA
- CONOPS 1: man portable tool
- CONOPS 2: continuous unattended verification

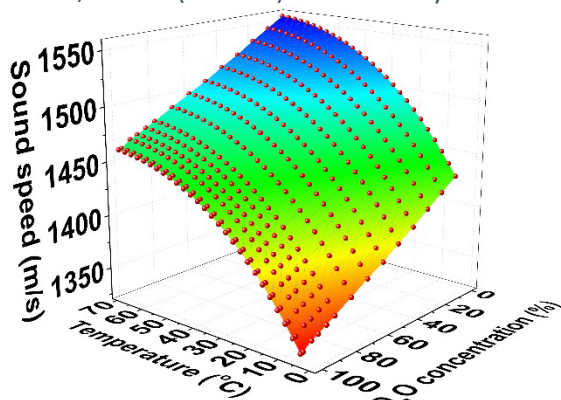
## Project Description

Determine accurate sound speed in fluid using Swept Frequency Acoustic Interferometry (SFAI).

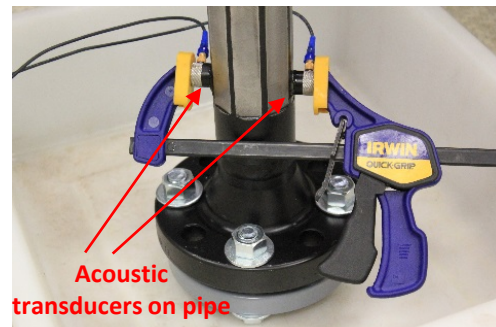
The approach consists of a clamp-on type device that can accurately measure D<sub>2</sub>O concentration in real-time in real settings.

## Current Capabilities (TRL-5)

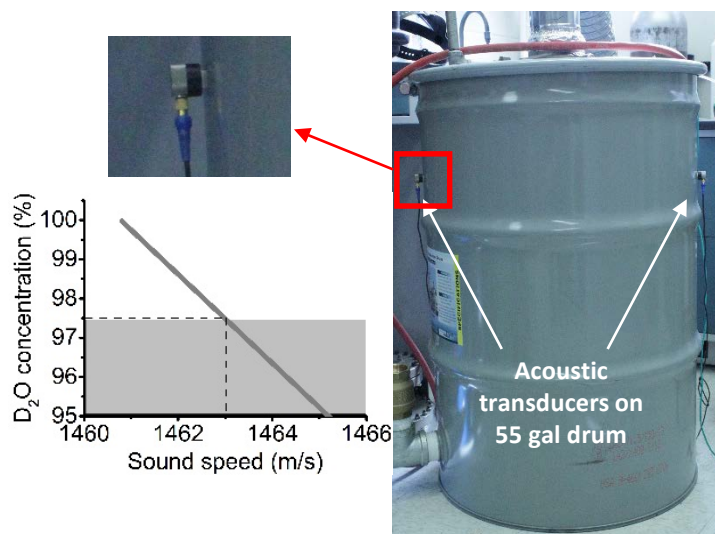
We already demonstrated high precision/accuracy for D<sub>2</sub>O concentration, ~ 0.1% (relative) in laboratory.



Graphical representation of sound speed, vs temperature and D<sub>2</sub>O concentration.



Example of acoustic transducers arrangements, for measurements on process pipes.



Example of acoustic transducers arrangements, for measurements on 55 gal drums.

## Anticipated Final Capabilities

We envision this work as a 3-year project. The work is divided in two different concepts of operations (CONOPS). One is a portable system for onsite inspection, while the other is a fixed dual-use system for unattended monitoring/verification.

### End of project :

Demonstration of continuous unattended tool in field.

## Further Reading

Sinha, D., Noninvasive determination of sound speed and attenuation in chemical warfare agents, in Handbook of Elastic Properties of Solids, Liquids and Gases, vol 4, p 3-21. 2001: Academic.